

ACCEPTED FOR PUBLICATION by
ENVIRONMENT INTERNATIONAL

**Determination of Volatile Organic Compounds and ETS Apportionment
in 49 Homes**

David L. Heavner, Walter T. Morgan, and Michael W. Ogden

R. J. Reynolds Tobacco Company, Research and Development, Winston-Salem,
North Carolina, 27102-1487

Address correspondence to:

David L. Heavner
Research and Development
R. J. Reynolds Tobacco Company
P.O. Box 1487
Winston-Salem, NC 27102-1487

Tel: 910-741-4982
FAX: 910-741-0719

Forty-nine nonsmoking married women participated in a home personal exposure study for 28 target volatile organic compounds (VOCs) and total volatile organic compounds (TVOCs). The women were selected and classified according to 18 socioeconomic categories based on age (18-34, 35-49, 50-64), family income (<\$25K, \$25K-\$40K, >\$40K), and husband's smoking status. Of the 29 analytes, 21 demonstrate no statistically significant difference in exposure between nonsmoking and smoking homes. One VOC, trichloroethylene, is elevated in the nonsmoking homes and seven VOCs, benzene, styrene, pyridine, 2-picoline, 3-picoline, 3-ethylpyridine, and 3-ethenylpyridine, are elevated in the smoking homes. A correlation matrix and a factor analysis indicate that benzene and styrene are not significantly correlated or associated with 3-ethenylpyridine, a proposed vapor phase environmental tobacco smoke (ETS) marker. All of the nitrogenous bases are significantly correlated with 3-ethenylpyridine. Benzene, styrene, and TVOC are not significantly correlated with the number of cigarettes smoked; however, 3-ethenylpyridine is significantly correlated with the number of cigarettes smoked. A Pearson correlation analysis indicates that "gas heat" and "smoking husband" are significantly correlated with elevated benzene concentrations, but a multiple regression model for benzene accounts for less than 30% of the total variance. ETS variables account for only 8% of the total variance. In the smoking homes, an apportionment technique is evaluated for selected VOCs in order to determine the median percentage of each analyte attributable to ETS. The results, with percentages:

attributable to ETS are: TVOC (5.5%), benzene (13.2%), styrene (12.6%), pyridine (40.7%), 2-picoline (67.1%), 3-picoline (90.1%), 4-picoline (37.2%), and 3-ethylpyridine (62.0%). Indoor air sources other than ETS are also identified for limonene, tetrachloroethylene, 1,4-dichlorobenzene, and alkylbenzenes.

Introduction

Studies on human exposure to target volatile organic compounds (VOCs) have increased dramatically in recent years. A few of the studies concentrate on outdoor aliphatic and aromatic VOC exposure while driving automobiles (Adlkofer et al. 1990; Chan et al. 1991; Dasch and Williams 1991), riding bicycles on urban and suburban thoroughfares (Bevan et al. 1991), or walking on sidewalks near busy city streets (Chan et al. 1991), but most of the studies focus on measurements in indoor air environments (Adlkofer et al. 1990; Bayer and Black 1986; Bayer and Black 1987; Brown et al. 1990; Chan et al. 1990; Girman et al. 1986; Higgins et al. 1987; McKone and Knezovich 1991; Michael et al. 1990; Miksch et al. 1982; Mølhave 1982; Mølhave 1990; Proctor 1988; Proctor 1989; Proctor et al. 1989; Proctor et al. 1991; Wallace 1986a; Wallace 1986b; Wallace 1987; Wallace et al. 1987; Weschler et al. 1990). Indoor VOC concentrations are generally higher than outdoor concentrations, and since people tend to spend most of their time indoors at home or in the workplace, the potential for VOC exposure is greatest in indoor settings (Wallace 1987a). Although simple quantitation of target VOCs is meaningful, source characterization and apportionment are vital and integral

parts of any indoor air evaluation. Numerous sources of indoor air VOCs exist: unvented kerosene or oil heaters, gas or oil furnaces, gas stoves, consumer products such as air fresheners, moth balls, and toilet bowl deodorizers, building or furnishing materials, clothing, use of hot water, and infiltration of VOCs from outdoor sources. However, one of the most controversial and publicized sources of indoor air VOCs is environmental tobacco smoke (ETS). The VOCs contributed by ETS constitute a heretofore undetermined fraction of a complex, multisource mixture of indoor air volatile compounds.

Tobacco does contribute to VOC levels in indoor environments by the very nature of cigarette use; that is, sidestream smoke and exhaled mainstream smoke generated from cigarettes as a result of combustion or pyrolysis mechanisms consequently emit VOCs into the surrounding environment. In a recent study (Heavner et al. 1992), a technique of ETS apportionment was proposed to assess the effect of smoking activity on specific VOC concentrations. The technique is based on the ratio of 3-ethenylpyridine, a vapor phase ETS marker, to other VOCs found in ETS.

The purpose of this investigation is to conduct a personal VOC exposure study of "smoking" and "nonsmoking" homes in a demographically selected U.S. city without the confounding effect of workplace exposure and to (1) determine the concentration of TVOCs (Wallace et al. 1991) and 28 target VOCs in these homes; (2) compare the VOC and TVOC levels in smoking and nonsmoking homes; (3) develop a correlation matrix to

compare VOC and TVOC concentrations with respect to each other; (4) assess the effect of specific lifestyle activities on selected VOC concentrations; (5) evaluate the relationship between the proposed vapor phase ETS marker, 3-ethenylpyridine, and other indicators of smoking activity such as indoor air nicotine and salivary cotinine concentrations; and, most importantly, (6) evaluate the ETS apportionment technique in order to determine the contribution of ETS to TVOC and selected target VOC concentrations in indoor air environments.

Experimental Methods

Apparatus, Materials and Chemicals. Personal VOC multisorbent samplers consisted of two stainless steel cartridges connected in series and packed with 160 mg of 60/80 mesh Tenax TA (Alltech Associates, Deerfield, IL) and 160 mg of 60/80 mesh Carbotrap (Supelco Inc., Bellefonte, PA) in each cartridge. The solid adsorbent sample cartridges were connected to Model 222-4 "low-flow" diaphragm pumps (SKC South, Appomattox, VA) with approximately 30 cm. of silicone rubber tubing. A pen clip was connected onto the front cartridge to facilitate attachment of the sampler cartridges to a lapel, collar, or pocket in the breathing zone of the participant. Pump flows (60-80 mL/min) were measured before and after the sample period, and the average of the two flows was used to calculate the VOC concentrations. Sample cartridges were analyzed by thermal desorption/gas chromatography/mass spectrometry for TVOCs and 28 target VOCs. Collection efficiency correction factors for each VOC were calculated and applied in

order to determine the amount entering the front cartridge, while perflubron concentrations were obtained by summing the mass determined on the front and back cartridges. In addition, ten cartridges were used for field blank correction. Data were analyzed statistically with SAS[®] software (SAS Institute Inc., Cary, NC). Sampler construction, sample collection, and analytical techniques are described in detail elsewhere (Heavner et al. 1992).

Diffusion monitors or passive sampling devices (PSDs) for five-day time-weighted average nicotine and 3-ethenylpyridine determinations were constructed from three-piece 37-mm polystyrene filter cassettes (Millipore Corp., Bedford, MA) with a Teflon membrane filter windscreen (Schleicher & Schuell, Keene, NH) and a glass fiber filter (Pallflex Co., Putnam, CT) treated with a 4% sodium bisulfate (Aldrich Chemical Co., St. Louis, MO) solution as the collection medium. Prior to, and immediately following sample collection, the PSDs were stored in polypropylene specimen jars with screw-cap lids (Fisher Scientific, Pittsburgh, PA). After sample preparation, filters were analyzed by gas chromatography/thermionic-specific (nitrogen) detection. PSD sampling rates for nicotine and 3-ethenylpyridine were 31.5 mL/min and 27.8 mL/min, respectively. PSD construction, validation, determination of sampling rates, sample preparation, and analytical techniques are described in detail elsewhere (Ogden and Maiolo 1992; Ogden et al. 1993).

Saliva samples were collected with Salivettes (Sarstedt Inc., Newton, NC). The devices

consisted of clear polystyrene conical centrifuge tubes with a snap-on cap and an untreated cylindrical cotton swab. After saliva collection, the Salivettes were placed in individual envelopes and stored at approximately -10°C in a freezer prior to cotinine determination by radioimmunoassay (RIA). The RIA method is described in detail elsewhere (Langone 1973; Langone and Van Vunakis 1982).

All sample media (VOC samplers, PSDs, and Salivettes) were collected from the participants over a five-day period and stored at approximately -10°C in freezers at a central location. After all sample collection materials were received, the media were packed with dry ice and shipped to the laboratory where they were immediately transferred to freezers and stored until analysis.

Site Selection and Timing. A national market research agency was contracted to provide guidance and assistance with regard to selection of a site city and study dates, development of a questionnaire and diary, participant training, and general administrative aspects of the VOC project. A local field service in the site city was subcontracted to provide a central facility for participant recruitment/training and collection of sample materials. Also, the national market research agency provided expertise as a liaison between the local field service, study participants, and client technical personnel involved in the study to ensure that neither the participants nor the local field service personnel were aware of the purpose of the study or the company sponsoring the project.

Columbus, Ohio during the winter week of February 25-29, 1991 was selected as a suitable site and time. Columbus is described by the American Automobile Association (AAA) as "one of the most average cities in the country" because its residents comprise a "demographically normal" segment of the U.S. population (Illinois/Indiana/Ohio Tourbook 1991). Also, the AAA states that Columbus offers a "perfect cross section of consumers" and is often referred to as "Test Market, U.S.A." Columbus has a population of approximately 560,000 with a metropolitan area population of over one million residents. In addition, Columbus is known as a center for scientific and technical information, retail banking, insurance, and real estate without the traditional heavy industrial base found in many midwestern U.S. cities.

Questionnaire and Diary Development. Questionnaires and diaries were developed in a joint effort with the national market research agency, client researchers, and a client statistician to ensure that questions were concise and unambiguous for participants in all socioeconomic categories. Questions related to lifestyle, leisure activities, diet, etc. were included and arranged to conceal the fact that the major independent variable was the husband's smoking status. The following questionnaires, diaries, and instructional materials were developed as a result of these efforts: (1) a "Screening Questionnaire" form used for initial telephone recruitment of participants; (2) a "First Visit Survey" form used by participants upon entering the site market research agency facility; (3) a "Pump and Special Diary Instructions" form used by participants as instructional reference for VOC sampler/pump operation and diary completion; (4) a "Special Diary and Additional

Questions" form used to record activities during the VOC sample period; (5) an "Air Monitor: Instructions for Participant" form used to instruct and record information regarding the use of personal and fixed area PSDs in the homes; (6) a "Saliva Sampling" form used to instruct and record information regarding collection of saliva samples; (7) a "Monitor/Salivette Return Probe" form used by interviewers to question participants regarding placement, usage, and collection of PSDs and saliva samples; (8) a "Pump Return Probe" form used to question participants regarding operation of the VOC sampler/pump; and (9) a "Last Visit Survey" form used to question participants regarding hobbies, diet, use of consumer products, etc.

Each prospective married female participant was placed in one of 18 cell categories based on the following variables: the husband's smoking status (smoking or nonsmoking), the participant's age (18-34, 35-49, or 50-64 years old), and family income range (<\$25K, \$25-\$40K, or >\$40K per year). Homes that contained smoking household members other than the husband were rejected. Fifty-five participants were recruited by telephone from the local field service's in-house database for participation in the project representing 24 of the 57 residential area Zip Codes in the greater Columbus area. All 18 cell categories were filled with at least one participant in each cell.

Participant Training. Experienced interviewers from the local field service trained all female participants. Each participant was given two kits: a VOC kit and a PSD/saliva kit. The VOC kit contained instructional materials, an activity diary, a pre-calibrated

The participants were provided with a small apron in which to place the pump during the sampling period. The instruction form included a diagram of the sampler/pump and information regarding pump operation. The interviewer demonstrated the proper technique for wearing the VOC sampler with the pump in the apron and the multisorbent sampler cartridges clipped onto a lapel or collar. The procedures for turning the pump on and off, recording the start and stop times, and completing the "Special Diary and Additional Questions" forms were conveyed as well.

VOC Sampling. The participants wore the VOC sampler for three consecutive evening hours on either Monday, Tuesday, or Wednesday during the week of February 25-29, 1991 while both the husband and wife were at home. In addition, the participants completed a special activity diary segmented into 15-minute intervals over the three-hour sampling period. The female participant recorded activities pertaining to everyone in the household. On the day after collecting the three-hour VOC sample, the participants returned the sampler to the facility. The recorded pump start and stop times were verified, and the pump flow was measured for comparison to the initial pump calibration. Finally, the participants signed a form at the end of the study certifying that they operated the pump for three consecutive hours and recorded activities that occurred during that period.

Nicotine and 3-Ethenylpyridine PSD Sampling. All participants wore a personal PSD for five consecutive days. In the smokers' homes only, a PSD was placed for five

consecutive days in a light use area of the home where the husband smoked the least amount of cigarettes, e.g. a child's bedroom, and a heavy use area of the home where the husband and wife spent most of their time together, e.g. a family room or den. The participants were told that the PSD would detect odors and chemicals; the participants were not told that the PSD was specific for nicotine or 3-ethenylpyridine. Daily diaries were provided for each of the five days, Monday through Friday, February 25-29, 1991. Daily diaries were segmented into one-hour periods for recording activities that occurred. During that hour, the participants were asked to record the number of cigarettes that were smoked in their presence, the number of times they smelled exhaust fumes, and the number of times that other odors were noticed. Each participant was given the appropriate number of PSDs, capped and placed in screw-cap jars, to open on the morning of the first day of the study (Monday). On Friday, the participants were asked to place the PSDs in the jars, record the time at the end of the study, and return the materials to the market research facility. Upon returning to the facility with the PSDs and saliva samples on the last day of the study, the participants were questioned regarding use of the sample materials.

Saliva Sampling. Also included in the PSD/saliva kit were the saliva sample instructional materials, four Salivette tubes, a thermally insulated bag, a refreezable ice pack (Stanbel Ice-Pak Inc., Montreal, PQ), and four insulated envelopes. To illustrate proper use of the saliva collection system during the first visit, the participants were asked to remove one of the Salivettes, place the cotton swab in their mouth, and chew

vigorously for one minute. The cotton swab was returned to the tube, and the tube was inserted into one of the envelopes. The envelopes were collected and immediately placed in a freezer for a baseline cotinine determination. These samples were labelled as "Demo" for administrative purposes. The participants were instructed to collect similar saliva samples on Monday morning, Wednesday evening, and Friday evening. The participants were instructed to place all saliva samples in individual envelopes and to store the envelopes in a freezer immediately. On Friday, February 29, the participants were asked to place the frozen ice pack in the thermal bag with the three saliva samples and return the bag to the market research facility.

Results and Discussion

Participant Rejection Criteria. Fifty-five married females participated in the sample collection portion of the Columbus study: 27 married to nonsmokers and 28 married to smokers. However, based on salivary cotinine results greater than 10ng/mL, five women were eliminated from the data analysis portion of the study (Etzel 1990). Another woman married to a smoker was eliminated from the data analysis portion of the study when it was discovered that the room in which she and her husband spent the majority of their time was equipped with an air cleaning system and that her husband smoked in a separate room of the house. Inclusion of this participant's results in the smoking home category would potentially bias the results. As a result, 49 participants were included in the data analysis portion of the study: 24 women married to nonsmokers and 25 women

married to smokers.

VOC Summary Statistics. Summary statistics for all VOCs and TVOCs are reported in Table I. Results are separated into smoking and nonsmoking homes with the number of determinations for each analyte (n), the means, standard deviations, medians, minima, maxima, and Wilcoxon Rank Sum Test p values for determining statistically significant differences between smoking and nonsmoking homes. The Wilcoxon Rank Sum Test was used because the data are generally not normally distributed. Of the 29 analytes (TVOC and 28 VOCs), 21 demonstrate no significant difference between smoking and nonsmoking homes. One of the VOCs, trichloroethylene, is elevated in the nonsmoking homes. Seven of the VOCs are elevated in the smoking homes: benzene, styrene, pyridine, 2-picoline, 3-picoline, 3-ethylpyridine, and 3-ethenylpyridine (3-EP). Of these seven, four of the nitrogenous bases (pyridine, 2-picoline, 3-picoline, and 3-ethylpyridine) are generally considered to originate predominantly from tobacco sources, and, as a result, are elevated in the smoking homes. However, pyridine and many of the pyridine derivatives have been found at trace levels in foods such as fish, meat, vegetables, cereals, dairy products, and alcoholic beverages (Jori et al. 1983; Vernin 1982). In addition, pyridine is eliminated from the human body through the breath, urine, feces, and skin (Jori et al. 1983), and a number of the pyridine derivatives are used as chemical intermediates in the production of adhesives, fibers, resins, and pesticides (Windholz 1976). Therefore, detection of some of these nitrogenous bases is expected in homes without smoking activity. The other nitrogenous base, 3-EP, is considered tobacco-

specific and is a proposed vapor phase ETS marker (Eatough et al. 1989; Heavner et al. 1992). Of course, 3-EP is elevated in the smoking homes, thus providing a measure of smoking activity. However, 3-EP is found in four of the 24 nonsmoking homes, though at extremely low concentrations. The mean 3-EP concentration in nonsmoking homes is $0.08 \mu\text{g}/\text{m}^3$, while the mean 3-EP concentration in smoking homes is $1.28 \mu\text{g}/\text{m}^3$; corresponding median concentrations are less than the limit of detection (LOD) and $0.95 \mu\text{g}/\text{m}^3$, respectively. The maximum 3-EP concentration in the nonsmoking homes is $0.57 \mu\text{g}/\text{m}^3$, while the maximum 3-EP concentration in the smoking homes is $5.58 \mu\text{g}/\text{m}^3$.

Correlation Matrix. A matrix of Pearson correlation coefficients is presented in Table II to illustrate statistically significant correlations between individual VOCs and, ultimately, VOC compound classes; correlation results from all homes ($n=49$), smoking and nonsmoking, are included in the matrix. Significant correlations at $p \leq 0.05$ are represented by filled circles (●) while correlations at $0.05 < p \leq 0.10$ are represented by open squares (□). The proposed ETS marker, 3-EP, is significantly correlated at $p \leq 0.05$ with the five nitrogenous bases: pyridine ($r=0.65$), 2-picoline ($r=0.85$), 3-picoline ($r=0.88$), 4-picoline ($r=0.55$), and 3-ethylpyridine ($r=0.64$). Also, limonene ($r=0.24$) is correlated at $0.05 < p \leq 0.10$ with the ETS marker. Although benzene is elevated in smoking homes, benzene is not correlated with 3-EP at either significance level. However, benzene is significantly correlated with 2-picoline ($r=0.34$) at $p \leq 0.05$ and 3-ethylpyridine ($r=0.26$) at $0.05 < p \leq 0.10$. In the smoking homes ($n=25$), benzene is not significantly correlated with either 3-EP or any of the nitrogenous bases. Thus, the

Correlation between benzene and "tobacco-related" nitrogenous bases in all smoking homes and non-smoking homes is insignificant. Instead, in all homes, benzene is significantly correlated with a number of other aromatic VOCs: toluene ($r=0.41$), ethylbenzene ($r=0.46$), *o*-xylene ($r=0.45$), *m*-xylene ($r=0.49$), *p*-xylene ($r=0.44$), *n*-propylbenzene ($r=0.36$), *n*-butylbenzene ($r=0.30$), 1,2,3-trimethylbenzene ($r=0.28$), and 1,3,5-trimethylbenzene ($r=0.35$) at $p \leq 0.05$, and styrene ($r=0.26$) at $0.05 < p \leq 0.10$. Correlation coefficients of styrene with other VOCs are similar to those of benzene. Styrene is correlated at $p \leq 0.05$ with ethylbenzene ($r=0.35$), *o*-xylene ($r=0.36$), *m*-xylene ($r=0.34$), *p*-xylene ($r=0.35$), pyridine ($r=0.30$), *n*-propylbenzene ($r=0.28$), 2-picoline ($r=0.36$), 3-picoline ($r=0.28$), 1,2,3-trimethylbenzene ($r=0.31$), and 1,3,5-trimethylbenzene ($r=0.29$). Styrene is not significantly correlated with 3-EP, the vapor phase ETS marker. As is the case with benzene, styrene in the smoking homes ($n=25$) is not significantly correlated with either 3-EP or any of the "tobacco-related" nitrogenous bases at $p \leq 0.05$ or $0.05 < p \leq 0.10$. In general, correlation results presented in Table II demonstrate that aromatic hydrocarbons are correlated with aromatic hydrocarbons, aliphatic hydrocarbons are correlated with aliphatic hydrocarbons, and nitrogenous bases are correlated with nitrogenous bases.

Factor Analysis. Since a visual inspection of the correlation matrix suggests that the VOCs might occur in correlated groups, a factor analysis was performed on all 28 VOCs and TVOC. Factor analysis is a variable-directed statistical technique used to identify or derive new variables or factors that provide a better understanding of the data (Chatfield

and Collins 1980). The purpose of performing a factor analysis is to examine the covariance structure of variables and to identify factors that are related to groups of variables. The interpretation of factors is a subjective process, generally aided by rotating the factors so that each factor is either highly correlated with a given variable or uncorrelated with it. Then the factor can be said to be representative of commonalities in the set of variables with which it is highly correlated and free of association from the set of variables with which it is not correlated. Since a number of major indoor air VOC sources are known to exist, it is reasonable to expect that many of the VOCs are generated from a smaller number of unique, unrelated sources that can be isolated as groups of interrelated variables in a factor analysis.

Factor analysis results for the 29 analytes are reported in Table III. Seven factors are identified with the variance explained by each factor listed above the VOCs grouped in that factor. The variance explained is closely related to the number of variables accounted for by each factor group. VOCs with a correlation coefficient of $r \geq 0.50$ with each factor are listed in Table III to show how the factor analysis grouped the VOCs. Factor one contains only aromatic hydrocarbons. Factor two contains mostly aliphatic hydrocarbons and TVOC with an overlap of one aromatic hydrocarbon, isopropylbenzene. Factor three contains only the "tobacco-related" nitrogenous bases including 3-EP, the vapor phase ETS marker. Factor four contains 1,4-dichlorobenzene and two *n*-alkanes (*n*-nonane, *n*-decane) with which it is significantly correlated in the correlation matrix (Table II). Factor five contains tetrachloroethylene and toluene;

tetrachloroethylene is significantly correlated with toluene and no other VOC (Table II). Factor six contains trichloroethylene and limonene, and factor seven contains styrene. In general, the factor analysis demonstrates the presence of three main groups incorporating 22 of the 29 analytes. These groups are characterized by compound class: the aliphatic VOCs, the aromatic VOCs, and the "tobacco-related" nitrogenous bases. The remaining seven analytes are distributed among four smaller, miscellaneous groups. The most significant finding is that benzene is grouped with the aromatic VOCs and not the "tobacco-related" nitrogenous bases. In addition, styrene is in a factor grouping by itself and is not grouped with the "tobacco-related" nitrogenous bases. If ETS were the sole or even the predominant indoor air source of benzene or styrene, then these two compounds would be expected to group with the "tobacco-related" VOCs.

Miscellaneous VOC Source Identification. The VOC target list of 28 compounds includes chemicals that originate from a number of diverse sources. In order to establish sources of these compounds, questionnaire responses were reviewed and specific activities or consumer products were selected as independent variables to determine statistically significant differences in the dependent variable responses, i.e. selected VOC concentrations. No attempt was made to identify sources for all of the target VOCs. Instead, a select group of four compounds or classes of compounds was chosen for the miscellaneous source identification: limonene, tetrachloroethylene, 1,4-dichlorobenzene, and the aromatic hydrocarbons. ETS as a source of specific VOCs will be addressed later in this discussion.

Limonene is a naturally-occurring compound found in the essential oils of a number of citrus fruits and herbs such as lemons, oranges, caraway, dill, and bergamot (Winholt 1976). Because of its lemon-like odor, it is often used in the consumer product industry as a fragrance to improve the sensory acceptability of products. In the "Last Visit Survey," participants were asked if they used certain consumer products on a fairly regular basis. Twelve consumer products were selected as potential sources of limonene: air freshener, liquid bleach, furniture polish, hair spray, kitchen cleaners, window cleaner, fingernail polish, carpet freshener, carpet cleaner, stain remover, perfume, and bug killer. Exposure to ETS during the three-hour "pump period" was selected as an additional independent variable for testing limonene concentrations since limonene is correlated with 3-EP at $0.05 < p \leq 0.10$ (Table II). In Table IV, Wilcoxon Rank Sum Test results are presented to compare limonene levels in the households where potential source products were used to households where they were not used, with significance indicated at $p \leq 0.05$. Results are tabulated separately for smoking/nonsmoking homes and all homes combined. In the nonsmoking homes, elevated limonene concentrations are associated with the use of hair spray or perfume ($p \leq 0.05$), and air freshener, furniture polish, and stain remover ($0.05 < p \leq 0.10$). In the smoking homes, elevated limonene concentrations are associated with the use of stain remover ($p \leq 0.05$). In all homes combined, elevated limonene concentrations are associated with the use of furniture polish and stain remover ($p \leq 0.05$ level) and hair spray ($0.05 < p \leq 0.10$). Exposure to ETS during the pump period appears to be statistically unrelated to limonene concentrations in the smoking homes even though limonene is correlated with 3-EP in

the study at 0.05 α p = 0.05.

Tetrachloroethylene is a known commercial dry-cleaning solvent, and is often used as a degreasing agent in commercial organic solvent-based cleaners (Windholz 1976).

Trichloroethylene has been used in the past as a commercial dry-cleaning solvent; however, current applications include use as a degreasing agent and as a solvent for numerous chemical preparations and coatings, such as paints and varnishes (Juhola 1973; Windholz 1976). In Table V(a), Wilcoxon Rank Sum Test results are presented to compare levels of trichloroethylene and tetrachloroethylene for groups of households defined by the activity of wearing clothes that were dry-cleaned within the previous week. Results are tabulated separately for smoking and nonsmoking homes and all homes combined with significance indicated at $p \leq 0.05$. In the nonsmoking homes and smoking homes separately, neither trichloroethylene nor tetrachloroethylene concentrations are significantly elevated with the activity of wearing dry-cleaned clothes; however, when all homes are combined, elevated tetrachloroethylene concentrations are associated with the activity of wearing dry-cleaned clothes ($p \leq 0.05$). Statistical power for this test is low since only two of the 49 participants wore clothes that had been dry cleaned within the previous week.

1,4-Dichlorobenzene is a volatile, crystalline solid used as an insecticidal fumigant in domestic mothball preparations, air fresheners/deodorizers, and toilet bowl deodorizers (Wallace 1987a; Windholz 1976). In Table V(b), Wilcoxon Rank Sum Test results are

presented to compare levels of 1,4-dichlorobenzene for groups of households defined by the activity of using mothballs in the home. Results are tabulated separately for smoking and nonsmoking homes and all homes combined with significance indicated at $p \leq 0.05$. In the nonsmoking homes and smoking homes separately, 1,4-dichlorobenzene concentrations are not significantly correlated with the activity of using mothballs in the home; however, when all homes are combined, elevated 1,4-dichlorobenzene concentrations are associated with the activity of using mothballs ($p \leq 0.05$). The ability to detect a significant effect in all homes combined is apparently due to an increase in statistical power.

A typical commercial grade of gasoline is a mixture of C_4 to C_{12} hydrocarbons containing aromatics, paraffins, and olefins (Winholz 1976)). It is a volatile mixture that could potentially affect indoor VOC concentrations, if gasoline is stored in a basement or attached garage. In Table VI, Wilcoxon Rank Sum Test results are presented to compare levels of the aromatic hydrocarbons and households defined by the activity of storing gasoline in an attached garage or basement. Again, results are tabulated separately for the smoking and nonsmoking homes and all homes combined with significance indicated at $p \leq 0.05$. In the nonsmoking homes, *o*-xylene ($p \leq 0.05$), toluene, isopropylbenzene, and *n*-propylbenzene ($0.05 < p \leq 0.10$) concentrations are significantly elevated in homes where gasoline is stored. In the smoking homes, *p*-xylene, *o*-xylene, *n*-propylbenzene ($p \leq 0.05$), *m*-xylene, and 1,3,5-trimethylbenzene ($0.05 < p \leq 0.10$) concentrations are significantly elevated in homes where gasoline is stored. In all

homes combined; ethylbenzene; p-xylene; isopropylbenzene; o-xylene; n-propylbenzene; 1,3,5-trimethylbenzene; 1,2,3-trimethylbenzene ($p \leq 0.05$), and m-xylene ($0.05 < p \leq 0.10$) are significantly elevated in homes where gasoline is stored. It is somewhat surprising that benzene and toluene do not demonstrate significance at either level since benzene and toluene are known to exist in gasoline formulations and automobile exhaust (Robinson et al. 1988).

Three-Hour Versus Five-Day Smoking Activity. The three-hour VOC samples collected in each of the 49 homes provide no information related to activities and subsequent exposures that occur on a daily, weekly, or yearly basis. In essence, the VOC samples collected in the homes provide a three-hour "snapshot" of activities and VOC exposure; however, it is important to establish whether or not the three-hour sampling period is typical of, or at least related to, longer-term smoking activity and VOC exposure. In Table VII, results are presented for a Pearson correlation analysis comparing the three-hour 3-EP concentrations with the five-day PSD nicotine, the five-day PSD 3-EP, and the salivary cotinine concentrations. Significant correlations at $p \leq 0.05$ are demonstrated for all comparisons with the exception of the Monday morning salivary cotinine ($0.05 < p \leq 0.10$). In general, the correlation analysis results suggest that the three-hour VOC sampler 3-EP concentrations are typical of the week's smoking activity.

For 3-ethenylpyridine, the three-hour 3-EP concentrations are significantly correlated ($p \leq 0.05$) with the five-day 3-EP "light" PSD ($r=0.88$), the five-day 3-EP "heavy" PSD

($r=0.83$), and the five-day 3-EP average PSD ($r=0.89$) concentrations. Thus, a strong statistical relationship exists between the short-term (three hour) 3-EP and the long-term (five day) 3-EP concentrations. In contrast, although the three-hour 3-EP concentrations are significantly correlated with the five-day 3-EP personal PSD ($r=0.72$) concentrations, the relationship is somewhat weaker.

The three-hour VOC sampler 3-EP and the five-day PSD nicotine relationship is similar. For example, the three-hour 3-EP concentrations are significantly correlated ($p \leq 0.05$) with the five-day nicotine "light" PSD ($r=0.46$), the five-day nicotine "heavy" PSD ($r=0.84$), and the five-day nicotine average PSD ($r=0.78$) concentrations. Again, the direct comparison of short-term 3-EP with long-term nicotine demonstrates a strong, statistically significant relationship with the exception of the "light" monitor which has the smallest correlation coefficient. Given the decay properties of nicotine (Nelson et al 1990) and the location of the "light" monitor in the child's bedroom or other seldom used area distant from the majority of smoking activity, a weaker correlation is expected.

For salivary cotinine, the three-hour 3-EP concentrations are significantly correlated ($p \leq 0.05$) with the "Demo" sample ($r=0.61$), the Wednesday evening sample ($r=0.71$), the Friday evening sample ($r=0.64$), the average of all samples ($r=0.62$), and the Monday morning salivary cotinine ($r=0.36$) at $0.05 < p \leq 0.10$. Since cotinine has a half-life of approximately 20 hours in humans (Benowitz 1983), all of the salivary cotinine samples, with the exception of the Monday morning sample, are indicative of exposure that

occurred during the week; the Monday morning sample is indicative of exposure that occurred on a weekend. If one assumes that weekday activities differ from weekend activities, then the Monday morning saliva sample is less typical of exposures that occur during the week. Overall, the three-hour VOC sampler 3-EP concentrations are more strongly correlated with the five-day PSD determinations, 3-EP and nicotine, than with salivary cotinine concentrations. Since the metabolic response of individuals to nicotine exposure is highly variable (Idle 1990), it is not surprising that the relationship between salivary cotinine concentrations and VOC sampler 3-EP concentrations is not as strong as the relationship between the PSD concentrations and VOC sampler 3-EP concentrations.

Number of Cigarettes Smoked. Information on the number of cigarettes observed during the "pump period" was recorded on the "Special Diary and Additional Questions" form. In Table VIII, results from a Pearson correlation analysis comparing the number of cigarettes smoked with 3-EP, benzene, styrene, and TVOC concentrations are reported for the smoking homes. In addition to the "cigarettes observed" variable, two normalized variables, "cigarettes observed/room" and "cigarettes observed/room excluding bathrooms" are included. The normalized variables are included to account for differences in home size. Ideally, the normalization should be performed with the actual house volume and/or measured ventilation rates; however, these variables were neither measured nor estimated.

Of the four analytes, only 3-ethenylpyridine, the vapor phase ETS marker, exhibits a

statistically significant correlation with any of the smoking activity variables. The cigarettes observed variable ($r=0.29$), is not significantly correlated with 3-EP; however, the two normalized variables, cigarettes observed/room ($r=0.40$) and cigarettes observed/room excluding bathrooms ($r=0.40$), are significantly correlated at $p \leq 0.05$. Even though two of the three correlations are significant, the correlation coefficient, r , is small; however, several factors may affect this relationship: (1) room area variability; (2) house volume variability; (3) air exchange rate variability; (4) sink source variability; (5) counting or reporting errors; and (6) ETS cigarette delivery variability.

The remaining selected analytes, benzene ($r=0.018$), styrene ($r=-0.03$), and TVOC ($r=-0.04$), are not significantly correlated with the number of cigarettes observed at $p \leq 0.05$ or $0.05 < p \leq 0.10$. Benzene ($r=0.16$), styrene ($r=0.01$), and TVOC ($r=0.02$) are not significantly correlated with the number of cigarettes observed/room. In addition, benzene ($r=0.18$), styrene ($r=0.01$), and TVOC ($r=0.01$) are not significantly correlated with the number of cigarettes observed/room excluding bathrooms. Although benzene and styrene are elevated in smoking homes versus nonsmoking homes as determined by the Wilcoxon Rank Sum Test (Table I), apparently several other sources contribute significantly to this elevation. In fact, benzene, styrene, and TVOC concentrations exhibit poorer correlation coefficients and p values with the normalized smoking activity variables than with the raw smoking activity variable, whereas the normalized smoking activity variables improve the correlation coefficients and p values with respect to 3-EP.

Benzene Source Identification. In order to evaluate benzene sources, a number of potential indoor and outdoor sources of benzene were identified and selected from questionnaire responses: smoking husband, gas or oil heat, fireplace, gas jets (logs), wood stove, gas range, cooking dinner, kerosene heater, type of home (apartment or single-family dwelling), attached garage, storing chemicals in the home, and distance from a highway. Benzene contributions from each of these potential sources were evaluated by conducting a Wilcoxon Rank Sum Test on the levels of benzene observed in homes where a given source was present, compared to homes in which it was not present. A Kruskal-Wallis Test was conducted for six levels of the distance from highway variable. The p values for these tests are given in Table IX. Only smoking husband, gas heat, gas or oil heat, and electric heat sources are significant at $p \leq 0.05$. Of the 49 homes, 38 used gas heat, 9 used electric heat, one used oil heat, and one reported geothermal heat. Thus, the set of homes heated with gas or oil is nearly identical to those heated with gas, and both are approximately the complement of those that used electricity. It should be noted that only one home reported the use of gas jets (logs), a woodstove, or a kerosene heater, and only two reported the use of a fireplace during the pump period. Therefore, tests of these potential sources have very little statistical power. Of the 17 potential sources, only smoking husband and gas vs. electric heat are sources that result in significantly different levels of benzene.

Benzene Regression Model. Using the dichotomous variables addressed in the previous section (Table IX) and the continuous variables related to the number of cigarettes

smoked during the three-hour period, a regression model was developed in an attempt to account for the variables resulting in elevated benzene concentrations. In Table X, results of a Pearson correlation analysis for the continuous variables and a benzene regression analysis are presented. For purposes of discussion, the variables are classified as either "ETS" or "non-ETS" variables. For the non-ETS variables, the regression model explains 23% of the total variance encountered; however, the overall model is insignificant ($p=0.73$). Adding the dichotomous ETS variable, smoking husband, to the non-ETS variables improves the model slightly by explaining 28% of the total variance; however, the overall model remains insignificant ($p=0.61$). The incremental addition of the smoking husband variable to the model is also insignificant at $p=0.15$. Removing this variable and adding the continuous ETS variable, cigarettes observed/room excluding bathrooms, to the non-ETS variables improves the model slightly by explaining 27% of the total variance; however, the overall model remains insignificant ($p=0.66$). The incremental addition of cigarettes observed/room excluding bathrooms is also insignificant at $p=0.21$. Removing this variable and adding the continuous ETS variable, cigarettes observed/room, to the non-ETS variables improves the model slightly by explaining 26% of the total variance; however, the overall model remains insignificant ($p=0.68$). The incremental addition of cigarettes observed/room is also insignificant at $p=0.22$. Removing this variable and adding the continuous ETS variable, cigarettes observed, to the non-ETS variables improves the model slightly by explaining 27% of the total variance; however, the overall model remains insignificant ($p=0.68$). The incremental addition of cigarettes observed is also insignificant at $p=0.24$. For the dichotomous and

continuous ETS variable alone, the regression model explains 8% of the total variance encountered; however, the overall model is insignificant ($p=0.42$). Finally adding all of the ETS and non-ETS variables to the model explains 30% of the total variance encountered; again, the overall model is insignificant ($p=0.78$). In conclusion, neither the ETS nor the non-ETS variables separately or combined yield significant model regression coefficients with respect to the benzene contributions. The non-ETS variables account for more of the variance ($r^2=0.23$) than the ETS variables ($r^2=0.08$). Obviously, the model's ability to identify and account for benzene sources other than gas heat and smoking husbands is weak. Apparently, the sum total of all other unidentified benzene sources exerts an overall greater effect on benzene concentrations than the variables applied in the model.

ETS Apportionment. The statistical techniques described thus far such as ranking, correlation analysis, factor analysis, and regression modeling are limited in their ability to determine true apportionment of VOC sources. For example, absolute benzene and styrene concentrations are higher in the 25 smoking homes than in the 24 nonsmoking homes, but correlation analysis and factor analysis methods indicate that the benzene and styrene concentrations are not correlated with the ETS marker concentrations (Tables II and III). In addition, benzene and styrene concentrations are not correlated with the other smoking-related indicators such as the number of cigarettes observed during the sample period (Table VIII). Finally, the benzene regression model is unable to predict benzene level with reasonable confidence given the input source variables. In a situation

source in particular VOCs such as benzene is generated from multiple, unaccounted-for sources. Apportionment technique is needed to identify individual contributions to the total analyte concentration. Daisey et al. (1991) presented a technique using model estimates of the contribution of ETS to VOCs and suggested the possibility of an apportionment technique using "gas-phase nicotine or some other unique tracer of ETS and the ratio of nicotine (or tracer) to VOC in ETS." In an earlier study (Heavner et al. 1992), an apportionment technique was described in which the ratio of a vapor phase ETS marker, 3-ethenylpyridine, to selected analytes was determined in an environmental chamber across a range of air exchange rates (0 to 2 per hour) with ETS generated from a 1R4F test cigarette (University of Kentucky, Lexington, KY) as the sole source of VOCs. Then, the percentage of the VOC attributable to ETS ($\% \text{Analyte}_{\text{ETS}}$) was calculated based on the 3-EP/Analyte ratio from field determinations. The utility of this technique was tested in four smoking and four nonsmoking homes. (Heavner et al. 1992)

In Table XI, apportionment results from each of the 25 smoking homes with summary statistics are presented for eight selected analytes: TVOC, benzene, styrene, pyridine, 2-picoline, 3-picoline, 4-picoline, and 3-ethylpyridine. Because the data are not normally distributed, the medians are more meaningful than the means in comparing results. For TVOC, the median percentage attributable to ETS is 5.5%, ranging from 0.0% to 25.8%. For benzene, the median percentage attributable to ETS is 13.2%, ranging from 0.0% to 63.2%. For styrene, the median percentage attributable to ETS is 12.6%, ranging from 0.0% to 58.1%. For the nitrogenous bases, pyridine, 2-picoline, 3-picoline, 4-picoline, and

ethylpyridine, the median percentages attributable to ETS are 40.7%, 67.1%, 90.1%, 57.2%, and 62.0%, respectively. For all of the nitrogenous bases, percentages attributable to ETS are greater than 100% in some of the homes. In these homes, the absolute concentrations (in $\mu\text{g}/\text{m}^3$) are extremely low compared to the concentrations used to determine the 3-EP/Analyte ratios in the environmental chamber. Consequently, small differences in the field determinations may result in apportionment percentages greater than 100%. Rather than arbitrarily limiting these values at 100%, the uncorrected values are presented to illustrate the difficulty in determining apportionment percentages when the absolute concentrations approach the limit of quantitation for a specific analyte. Overall, the ETS apportionment technique demonstrates the multisource nature of VOCs in indoor air environments and provides one quantitative estimate of the percentage of selected VOCs that originate from smoking activity.

Furthermore, the %Analyte_{ETS} determinations may be used to calculate the absolute concentration of analytes originating from ETS and from all other sources combined. In Figure 1, the "Total" concentrations and the "ETS" concentrations are presented in a histogram format for four of the analytes (TVOC, benzene, styrene, and 3-picoline) from the 24 nonsmoking and the 25 smoking homes. The results are paired and ranked in increasing order of "Total" concentration. TVOCs are not calculated for three of the 49 homes due to GC/MSD instrument problems resulting in the loss of data from the back cartridges. The histograms graphically illustrate the ETS contribution to the total concentration and place into perspective the relationship between the portion originating

from ETS and the portion originating from other, non-ETS sources. In general, these results demonstrate that even in the absence of smoking activity, substantial levels of TVOCs and VOCs are generated. ETS does contribute to the total concentration of specific VOCs; however, the elimination of ETS as a source has minimal impact on the total concentration with the exception of the nitrogenous bases. Most importantly, ETS contributes relatively little to the aggregate VOC indicator, the TVOC concentration.

Conclusions

In this study, the levels of VOCs measured during a three-hour period in 25 smoking homes and 24 nonsmoking homes have been discussed and related to a number of multiple indoor air sources. Of the 29 analytes, one VOC is elevated in the nonsmoking homes, seven VOCs are elevated in the smoking homes, and 21 VOCs are not statistically different. Storing gasoline in a garage or basement contributes to elevated levels of eight aromatic hydrocarbons. Using mothballs in the home contributes to elevated levels of 1,4-dichlorobenzene. Wearing dry-cleaned clothes contributes to elevated levels of tetrachloroethylene. Using stain remover or furniture polish contributes to elevated levels of limonene. Although benzene and styrene are elevated in smoking homes, these VOCs are not significantly correlated with the number of cigarettes smoked or the proposed vapor phase ETS marker, 3-ethenylpyridine, indicating that a number of other unidentified sources contribute to this elevation. Based on nonparametric tests of 17 source input variables, gas heat and smoking husband are

identified as source variables contributing to indoor air benzene levels; however, a regression analysis model indicates that the ETS variables account for only 8% of the total variance encountered. The non-ETS variables account for 23% of the total variance encountered. The technique of ETS apportionment based on 3-ethenylpyridine/VOC ratios appears promising, but further investigation is required in order to determine 3-EP ratios for existing market cigarette brands. Nonetheless, this technique is potentially useful in assessing the portion of indoor air VOCs attributable to ETS in environments where multiple sources are responsible for contributing to the total concentration of a specific analyte. The median percentages of TVOC, benzene, and styrene attributable to ETS in smoking households are 5.5%, 13.2%, and 12.6%, respectively.

Acknowledgements

The authors would like to thank Mr. Robert B. Hege, Jr. for administrative assistance, Mr. Riley A. Davis and Mr. Mitchell E. Stiles for preparation and analysis of the saliva cotinine samples, and Ms. Katherine C. Maiolo for preparation and analysis of the PSD nicotine and 3-ethenylpyridine samples.

Literature Cited

Adlkofer, F.; Scherer, G.; Conze, C.; Angerer, J.; Lehnert, G. J. Significance of exposure to benzene and other toxic compounds through environmental tobacco smoke. *Cancer Res. Clin. Oncol.* 116:591-598; 1990.

Bayer, C.W.; Black, M.S. Passive smoking: survey analysis of office smoking areas vs. environmental chamber studies. *Proc. ASHRAE Conference IAQ'86*. Atlanta, GA; 1986:281-291.

Bayer, C.W.; Black, M.S. Thermal desorption/gas chromatographic/mass spectrometric analysis of volatile organic compounds in the offices of smokers and nonsmokers. *Biomed. Environ. Mass Spectrom.* 14:363-367; 1987.

Benowitz, N.L. The use of biologic fluid samples in assessing tobacco smoke consumption. In: *Measurement in the analysis and treatment of smoking behavior*. Washington, D.C.: U.S. Government Printing Office. NIDA Monograph 48: 6-25; 1983.

Bevan, M.A.J.; Proctor, C.J.; Baker-Rogers, J.; Warren, N.D. Exposure to carbon monoxide, respirable suspended particulates, and volatile organic compounds while

commuting by bicycle. Environ. Sci. Technol. 25:788-790; 1991.

Brown, V.M.; Crump, D.R.; Gardiner, D. Determination of aromatic hydrocarbon emissions from paint and related products by an impinger method. Environ. Int. 16:283-289; 1990.

Chan, C.C.; Özkaynak, H.; Spengler, J.D.; Sheldon, L. Driver exposure to volatile organic compounds, CO, ozone, and NO₂ under different driving conditions. Environ. Sci. Technol. 25:964-972; 1991.

Chan, C.C.; Vainer, L.; Martin, J.W.; Williams, D.T. Determination of organic contaminants in residential indoor air using an adsorption-thermal desorption technique. J. Air Waste Manage. Assoc. 40:62-67; 1990.

Chatfield, C.; Collins, A.J. Introduction to multivariate analysis. London, UK: Chapman and Hall Ltd.; 1980.

Daisey, J.M.; Gadgil, A.; Hodgson, A.T.; Model estimates of the contributions of environmental tobacco smoke to volatile organic compound exposures in office buildings. Indoor Air 1:37-45; 1991.

Dasch, J.M.; Williams, R.L. Benzene exhaust emissions from in-use General Motors

vehicles. Environ. Sci. Technol. 25:853-857; 1991.

Eatough, D.J. et al. The chemical composition of environmental tobacco smoke III. identification of conservative tracers of environmental tobacco smoke. Environ. Int. 15:19-28; 1989.

Etzel, R.A. A review of the use of saliva cotinine as a marker of tobacco smoke exposure. Preventive Med. 19:190-197; 1990.

Girman, J.R.; Hodgson, A.T.; Newton, A.S. Emissions of volatile organic compounds from adhesives with indoor applications. Environ. Int. 12:317-321; 1986.

Heavner, D.L.; Ogden, M.W.; Nelson, P.R. Multisorbent thermal desorption/gas chromatography/mass selective detection method for the determination of target volatile organic compounds in indoor air. Environ. Sci. Technol. 26:1737-1746; 1992.

Higgins, C.E.; Thompson, C.V.; Ilgner, R.H.; Jenkins, R.A.; Guerin, M.R. Multicomponent environmental tobacco smoke analysis using triple sorbent traps and thermal desorption gas chromatography. Presented 41st Tobacco Chemists' Research Conference. Greensboro, NC; 1987.

Illinois/Indiana/Ohio TourBook. Heathrow, FL: American Automobile Association; 1991.

Jori, A. et al. Ecotoxicological profile of pyridine: working party on ecotoxicological profiles of chemicals. *Ecotoxicol. Environ. Safety* 7:251-275; 1983.

Juhola, A.J. MSA Research. Package sorption device system study. USEPA Contract No. EHSD 71-2 EPA-R2-73-202; 1973.

Langone, J.J.; Gjika, H.B.; Van Vunakis, H. Nicotine and its metabolites: Radiomunoassays for nicotine and cotinine. *Biochemistry* 12:5025-5030; 1973.

Langone, J.J.; Van Vunakis, H. Radioimmunoassay of nicotine, cotinine, and γ -(3-pyridyl)- γ -oxo-N-methylbutyramide. In: *Immunochemical techniques; methods in enzymology*. New York, NY: Academic Press; 1982.

McKone, T.E.; Knezovich, J.P. The transfer of trichloroethylene (TCE) from a shower to indoor air: experimental measurements and their implications. *J. Air Waste Manag. Assoc.* 40:282-286; 1991.

... ..
... ..
... ..
for assessing personal exposure to volatile organic compounds. Environ. Sci.
Technol. 24:996-1003; 1990.

Milksch, R.R.; Hollowell, C.D.; Schmidt, H.E. Trace organic chemical contaminants in
office spaces. Environ. Int. 8:129-137; 1982.

Mølhave, L. Indoor air pollution due to organic gases and vapours of solvents in building
materials. Environ. Int. 8:117-127; 1982.

Mølhave, L. Volatile organic compounds, indoor air quality and health. Proc. 5th
International Conference on Indoor Air Quality and Climate. Vol. 5. Toronto,
Canada; 1990:15-33.

Nelson, P.R.; Heavner, D.L.; Oldaker, G.B. III. Problems with the use of nicotine as a
predictive environmental tobacco smoke marker. Proc. 1990 EPA/A&WMA
International Symposium: Measurement of Toxic and Related Air Pollutants.
Raleigh, NC; 1990:550-555.

Ogden, M.W.; Maiolo, K.C. Comparative evaluation of diffusive and active sampling
systems for determining airborne nicotine and 3-ethenylpyridine. Environ. Sci.

Journal of 123 123 1992.

Ogden, M.W. et al. Multiple measures of personal ETS exposure in a population-based survey of non-smoking women in Columbus, Ohio. Proc. 6th International Conference on Indoor Air Quality and Climate. Vol. 1. Helsinki, Finland; 1993:523-528.

Proctor, C.J. The analysis of the contribution of ETS to indoor air. In: Indoor and ambient air quality. London, UK: Selper Ltd.; 1988.

Proctor, C.J. A comparison of the volatile organic compounds present in the air of real-world environments with and without environmental tobacco smoke. Proc. Air and Waste Management Association Annual Meeting. Anaheim, CA; 1989:Paper 89-80.4.

Proctor, C.J.; Warren, N.D.; Bevan, M.A.J. Measurements of environmental tobacco smoke in an air-conditioned office building. Environ. Technol. Lett. 10:1003-1018; 1989.

Proctor, C.J.; Warren, N.D.; Bevan, M.A.J.; Baker-Rogers, J. A comparison of methods of assessing exposure to environmental tobacco smoke in non-smoking British women. Environ. Int. 17:287-297; 1991.

Robinson, M.R.; Bostwick, D.E.; Abbey, L.E.; Moran, T.F. Enhanced specificity for aromatics using 2E mass spectrometry. *Rapid Commun. Mass Spectrom.* 2(10):210-212; 1988.

Vernin, G. Heterocyclic compounds in flavors and fragrances: part III. pyridine and derivatives. *Perfum. Flav.* 7:23-26; 1982.

Wallace, L.A. Personal exposures, indoor and outdoor concentrations, and exhaled breath concentrations of selected volatile organic compounds measured for 600 residents of New Jersey, North Dakota, North Carolina and California. *Tox. Environ. Chem.* 12:215-236; 1986a.

Wallace, L.A. Cancer risks from organic chemicals in the home. *Proc. APCA International Specialty Conference*. Chicago, IL; 1986b:14-24.

Wallace, L.A. USEPA (U.S. Environmental Protection Agency). The total exposure assessment methodology (TEAM) study: summary and analysis: volume I. EPA-600/6-87/002a. Washington, D.C.: Office of Research and Development, USEPA; 1987.

Wallace, L.A.; Pellizzari, E.; Hartwell, T.D.; Perritt, R.; Ziegenfuss, R. Exposures to benzene and other volatile compounds from active and passive smoking. *Arch.*

Environ. Health 42:272-279; 1987.

Wallace, L.; Pellizzari, E.; Wendel, C.; Total volatile organic concentrations in
2700 personal, indoor, and outdoor air samples collected in the US EPA
TEAM studies. Indoor Air 4:465-477; 1991.

Weschler, C.J.; Shields, H.C.; Rainer, D. Concentrations of volatile organic compounds at
a building with health and comfort complaints. Am. Ind. Hyg. Assoc. J. 51:261-268;
1990.

Windholz, M., Ed. The Merck index. Rahway, NJ: Merck & Co., Inc.; 1976.

Table I. VOC summary statistics for smoking and nonsmoking homes.

Compound	Nonsmoking Homes ($\mu\text{g}/\text{m}^3$)						Smoking Homes ($\mu\text{g}/\text{m}^3$)						Wilcoxon
	n	Mean	S.D.	Median	Minimum	Maximum	n	Mean	S.D.	Median	Minimum	Maximum	
n-Nonane	24	3.00	2.60	2.01	0.51	8.95	25	3.84	4.18	2.41	0.54	16.67	0
Benzene	24	3.86	4.05	2.42	1.29	18.96	25	5.54	5.13	4.03	0.98	26.96	0
n-Decane	24	5.07	4.25	2.76	1.46	16.86	25	4.99	9.00	2.75	0.00	46.02	0
Trichloroethylene	24	1.84	2.39	1.03	0.00	9.08	25	0.66	1.04	0.00	0.00	3.41	0
Tetrachloroethylene	24	1.24	1.56	0.70	0.00	5.13	25	0.89	0.96	0.68	0.00	3.78	0
Toluene	24	19.25	13.11	13.65	3.01	47.43	25	27.78	22.17	23.79	4.42	118.20	0
1,2-Dichloroethane	24	0.00	0.00	0.00	0.00	0.00	25	0.02	0.08	0.00	0.00	0.40	0
n-Undecane	24	3.86	4.64	1.99	0.14	17.19	25	14.50	49.93	1.81	0.56	249.68	0
Ethylbenzene	24	3.35	4.87	2.12	0.36	25.39	25	3.07	3.57	2.21	0.82	19.45	0
p-Xylene	24	3.63	6.14	2.08	1.01	31.78	25	3.64	4.37	2.34	0.80	22.42	0
m-Xylene	24	7.24	9.30	4.45	1.02	47.21	25	7.57	7.58	5.71	0.16	35.96	0
Isopropylbenzene	24	0.46	3.05	0.26	0.00	5.11	25	0.45	0.66	0.27	0.00	3.23	0
o-Xylene	24	4.21	6.61	2.43	1.13	34.24	25	4.21	5.13	2.57	1.16	25.41	0
Pyridine	24	0.67	0.53	0.66	0.01	1.86	25	2.34	2.03	1.97	0.00	8.59	0
n-Dodecane	24	1.63	1.39	1.34	0.32	4.50	25	7.34	25.21	1.43	0.19	127.90	0
Limonene	24	17.81	11.27	13.82	3.45	41.98	25	23.15	15.94	20.77	5.68	57.39	0
n-Propylbenzene	24	1.06	1.91	0.60	0.23	9.88	25	0.93	1.26	0.52	0.00	5.20	0
2-Picoline	24	0.07	0.28	0.00	0.00	0.67	25	0.45	0.35	0.38	0.00	1.55	0
1,3,5-Trimethylbenzene	24	1.85	1.85	1.12	0.41	15.29	25	1.92	2.33	1.02	0.23	8.81	0
Styrene	24	1.47	1.02	1.30	0.43	4.96	25	2.11	1.20	1.92	0.49	7.02	0
n-Tridecane	24	1.41	0.55	1.28	0.38	2.73	25	2.67	3.95	1.26	0.28	15.59	0
3-Picoline	24	0.14	0.16	0.09	0.00	0.51	25	0.68	0.56	0.58	0.00	2.40	0
4-Picoline	24	0.09	0.19	0.00	0.00	0.69	25	0.16	0.26	0.00	0.00	0.94	0
n-Butylbenzene	24	0.27	0.36	0.16	0.00	1.39	25	0.34	0.41	0.26	0.00	1.54	0
1,2,3-Trimethylbenzene	24	1.76	2.10	1.29	0.37	10.91	25	2.19	2.69	1.18	0.42	10.11	0
3-Ethylpyridine	24	0.03	0.10	0.00	0.00	0.46	25	0.18	0.24	0.00	0.00	0.68	0
1,4-Dichlorobenzene	24	3.45	6.50	0.30	0.00	25.39	25	10.22	30.24	0.16	0.00	116.23	0
3-Ethethylpyridine	24	0.06	0.19	0.00	0.00	0.57	25	1.28	1.31	0.95	0.00	5.58	0
TVOC	22	289	251	191	56	921	24	458	282.5	252	79	14097	0

* Significant at $p \leq 0.05$

SENT BY:

: 8- 1-94 : 17:07 :

ETS/BGT-9

+49 221 1648 858: #41

2026224072

Table II. Pearson correlation matrix for VOC analytes in all homes (smoking and nonsmoking).

Compound	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)	(11)	(12)	(13)	(14)	(15)	(16)	(17)	(18)	(19)	(20)	(21)	(22)	(23)	(24)	(25)
(1) <i>n</i> -Nonane																									
(2) Benzene																									
(3) <i>n</i> -Decane																									
(4) Trichloroethylene																									
(5) Tetrachloroethylene																									
(6) Toluene																									
(7) 1,2-Dichlorobenzene																									
(8) <i>n</i> -Undecane																									
(9) Ethylbenzene																									
(10) <i>p</i> -Xylene																									
(11) <i>m</i> -Xylene																									
(12) Isopropylbenzene																									
(13) <i>o</i> -Xylene																									
(14) Pyridine																									
(15) <i>n</i> -Dodecane																									
(16) Linoleene																									
(17) <i>n</i> -Propylbenzene																									
(18) 2-Propanol																									
(19) 1,3,5-Trinitrobenzene																									
(20) Styrene																									
(21) <i>n</i> -Tridecane																									
(22) 3-Picoline																									
(23) 4-Picoline																									
(24) <i>n</i> -Butylbenzene																									
(25) 1,2,3-Trinitrobenzene																									
(26) 3-Ethylpyridine																									
(27) 1,4-Dichlorobenzene																									
(28) 3-Tertbutylpyridine																									
(29) TVOC																									

Significant at $p \leq 0.05$ ☐ Significant at $p \leq 0.01$ ☐ Significant at $p \leq 0.001$ ☐

Significant at $p \leq 0.05$ \square Significant at $p \leq 0.10$

2026224073

Table III. Factor analysis results for VOC analytes in all homes (smoking and nonsmoking).

	Correlation Between Variables and Factors ($r \geq 0.50$)						
	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Factor 7
Variance Explained by Each Factor	7.912	6.482	4.936	2.063	1.640	1.279	1.195
Compound & Correlation Coefficient	Benzene 0.61	n-Nonane 0.51	Pyridine 0.71	n-Nonane 0.68	Tetrachloroethylene 0.74	Trichloroethylene -0.53	Styrene 0.65
	Ethylbenzene 0.91	1,2-Dichloroethane 0.98	2-Picoline 0.92	n-Decane 0.91	Toluene 0.82	Limonene 0.73	
	p-Xylene 0.97	n-Undecane 0.97	3-Picoline 0.92	1,4-Dichlorobenzene 0.80			
	m-Xylene 0.98	Isopropylbenzene 0.59	4-Picoline 0.81				
	Isopropylbenzene 0.59	n-Dodecane 0.98	3-Ethylpyridine 0.84				
	o-Xylene 0.99	n-Tridecane 0.51	3-Ethylpyridine 0.89				
	n-Propylbenzene 0.98	TVOC 0.97					
	1,3,5-Trimethylbenzene 0.96						
	n-Butylbenzene 0.77						
	1,2,3-Trimethylbenzene 0.82						

SENT BY:

; 8-1-94 ; 17:13 ;

ETS/BGTC+

+49 221 1648 8581 #43

2026224074

SENT BY:

: 8- 1-94 : 17:15 :

ETS/BGTC-

+49 221 1648 858;#45

Table 1. Frequency of Use of Various Household Products by Smoking Status

Table 1 (continued)

Household Product	Wilcoxon Rank Sum Test		
	Nonsmoking Homes	Smoking Homes	All Homes
Air Freshener	0.08	0.62	0.58
Liquid Bleach	0.41	0.93	0.53
Furniture Polish	0.06	0.37	0.05*
Hair Spray	0.009*	0.92	0.06
Kitchen Cleaner	0.75	0.40	0.30
Window Cleaner	0.73	0.77	0.46
Fingernail Polish	0.41	0.68	0.75
Carpet Freshener	0.50	0.73	0.49
Carpet Cleaner	0.66	0.30	0.25
Stain Remover	0.10	0.05*	0.01*
Perfume	0.02*	0.39	0.21
Bug Killer	Not Used	0.15	0.19
Exposed to ETS During the "Pump Period"	---	0.83	0.32

* Significant at $p \leq 0.05$

2026224075

Table V. Wilcoxon Rank Sum Test results in all homes (smoking and nonsmoking): (a) trichloroethylene/tetrachloroethylene and wearing clothes that were dry-cleaned in the week prior to wearing the sampler; and (b) 1,4-dichlorobenzene and using mothballs in the home.

Compound	Wilcoxon Rank Sum p Value		
	Nonsmoking Homes	Smoking Homes	All Homes
(a) Trichloroethylene	0.66	0.14	0.18
Tetrachloroethylene	0.11	0.18	0.03*
(b) 1,4-Dichlorobenzene	0.20	0.12	0.03*

* Significant at p ≤ 0.05

2026224076

SENT BY:

; 8-1-94 ; 17:17 ;

ETS/BGTC→

+49 221 1648 858;#47

Comparison of indoor air concentrations of volatile organic compounds (VOCs) in nonsmoking and smoking homes.

	Nonsmoking Homes	Smoking Homes	All Homes
Benzene	0.88	0.48	0.70
Toluene	0.09	0.98	0.25
Ethylbenzene	0.12	0.18	0.05*
p-Xylene	0.17	0.04*	0.03*
m-Xylene	0.17	0.08	0.06
Isopropylbenzene	0.10	0.17	0.02*
o-Xylene	0.03*	0.03*	0.004*
n-Propylbenzene	0.08	0.05*	0.003*
1,3,5-Trimethylbenzene	0.12	0.10	0.01*
Styrene	0.90	0.89	0.40
n-Butylbenzene	0.21	0.23	0.12
1,2,3-Trimethylbenzene	0.13	0.20	0.04*

* Significant at $p \leq 0.05$

2026224077

	Correlation Coefficient (r)	p Value
5-Day Nicotine "Light" Use Area Fixed Monitor	0.46	0.0203*
5-Day Nicotine "Heavy" Use Area Fixed Monitor	0.84	0.0001*
5-Day Nicotine Average Fixed Monitors	0.78	0.0001*
5-Day Nicotine Personal Monitor	0.84	0.0001*
5-Day 3-Ethenylpyridine "Light" Use Area Fixed Monitor	0.88	0.0001*
5-Day 3-Ethenylpyridine "Heavy" Use Area Fixed Monitor	0.88	0.0001*
5-Day 3-Ethenylpyridine Average Fixed Monitors	0.89	0.0001*
5-Day 3-Ethenylpyridine Personal Monitor	0.72	0.0001*
"Demo" Salivary Cotinine	0.61	0.0012*
Monday Salivary Cotinine	0.36	0.0772
Wednesday Salivary Cotinine	0.71	0.0001*
Friday Salivary Cotinine	0.64	0.0006*
Average Salivary Cotinine	0.62	0.0009*
* Significant at $p \leq 0.05$		

2026224078

Table VIII. Pearson correlation analysis results comparing the number of cigarettes smoked with 3-ethenylpyridine, benzene, styrene, and concentrations in smoking homes.

Cigarettes	3-Ethenylpyridine		Benzene		Styrene		TVOC	
	Correlation Coefficient	P	Correlation Coefficient	P	Correlation Coefficient	P	Correlation Coefficient	P
Observed	0.29	0.15	0.18	0.39	-0.03	0.87	-0.04	0.85
Observed/Room	0.40	0.049*	0.16	0.45	0.01	0.96	0.02	0.92
Observed/Room Excluding Bathrooms	0.40	0.049*	0.18	0.40	0.01	0.97	0.01	0.97

* Significant at $p \leq 0.05$

2026224079

	n	Mean ($\mu\text{g}/\text{m}^3$)	Median ($\mu\text{g}/\text{m}^3$)	s	Mean ($\mu\text{g}/\text{m}^3$)	Median ($\mu\text{g}/\text{m}^3$)	
Smoking Husband	24	5.54	4.03	25	3.86	2.42	0.006*
Gas Heat	38	5.33	3.54	11	2.59	2.43	0.01*
Electric Heat	9	2.55	2.43	40	5.20	3.50	0.02*
Gas or Oil Heat	39	5.25	3.51	10	2.62	2.46	0.02*
Used Fireplace	2	5.91	5.91	47	4.66	3.22	0.18
Used Gas Jet	1	7.79	7.79	48	4.65	3.24	0.19
Used Wood Stove	1	1.81	1.81	48	4.78	3.36	0.19
Oil Heat	1	2.28	2.28	48	4.77	3.36	0.42
Distance from Highway [†]	—	—	—	—	—	—	0.45
Cooked Dinner	36	4.54	3.36	13	5.19	3.22	0.46
Gas Range	12	5.70	3.15	37	4.40	3.47	0.65
Used Kerosene Heater	1	2.72	2.72	48	4.76	3.36	0.70
Single Family Home	44	4.82	3.24	5	3.83	3.47	0.77
Store Chemicals in the Home	38	5.02	3.35	11	3.65	3.25	0.79
Attached Garage	35	5.11	3.25	14	3.72	3.26	0.79
Used Gas Range	8	6.25	3.15	41	4.42	3.47	0.86
Geothermal Heat	1	3.22	3.22	48	4.75	3.36	0.97

[†]Kruskal-Wallis Test (Six levels) * Significant at $p \leq 0.05$

2026224080

Table X. Pearson correlation analysis and regression model results comparing ETS and non-ETS sources with benzene concentration in all homes.

Source Variables Included in Regression Model	Pearson Correlation for "ETS" Variables (n=49)		Regression Analysis		
	Correlation Coefficient (r)	p Value	Model r^2	p Value Overall	p Value Incremental
All "Non-ETS" Variables	---	---	0.23	0.73	---
All "Non-ETS" Variables + Smoking Husband	0.18	0.21	0.28	0.61	0.15
All "Non-ETS" Variables + Cigarettes Observed/Room Exhausting Bathrooms	0.22	0.12	0.27	0.66	0.21
All "Non-ETS" Variables + Cigarettes Observed/Room	0.21	0.15	0.26	0.68	0.22
All "Non-ETS" Variables + Cigarettes Observed	0.22	0.12	0.27	0.67	0.24
All "Non-ETS" + All "ETS" Variables	---	---	0.30	0.78	---
All "ETS" Variables	---	---	0.08	0.42	0.57

2026224081

Table XL ETS apportionment results for selected analytes in smoking homes.

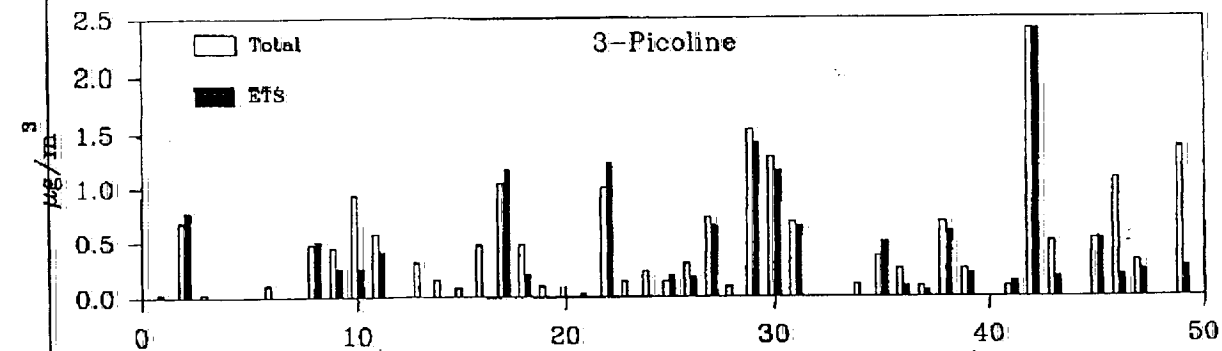
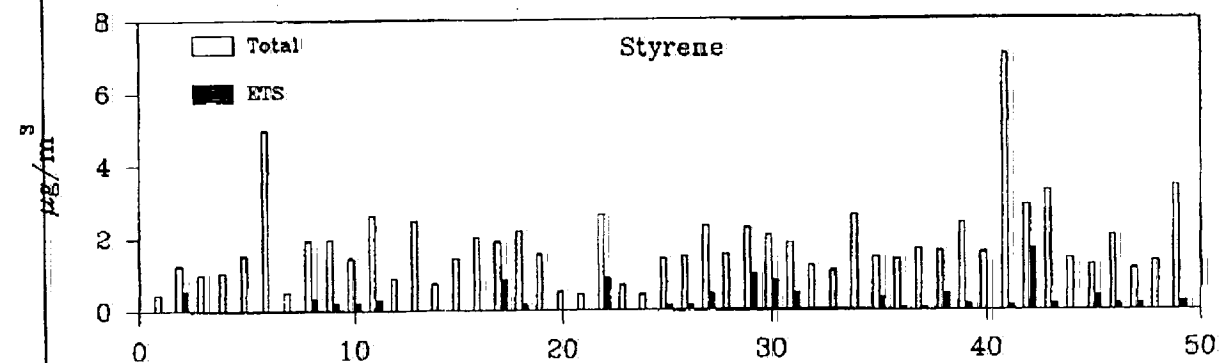
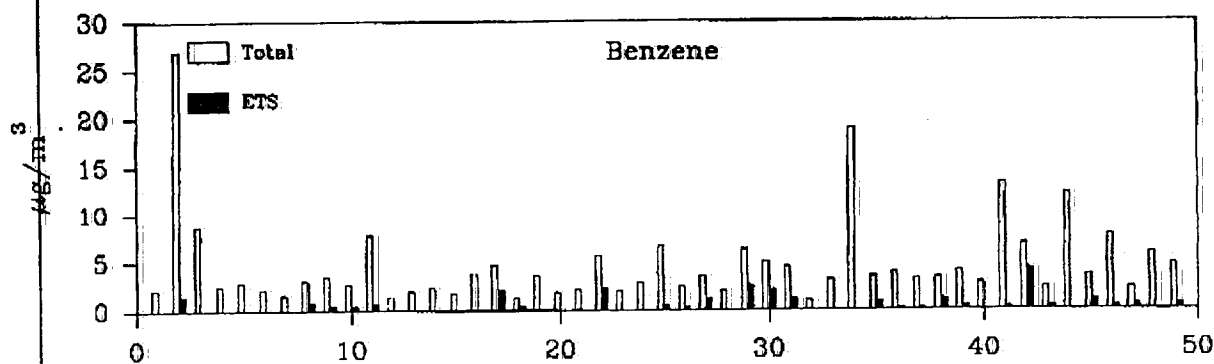
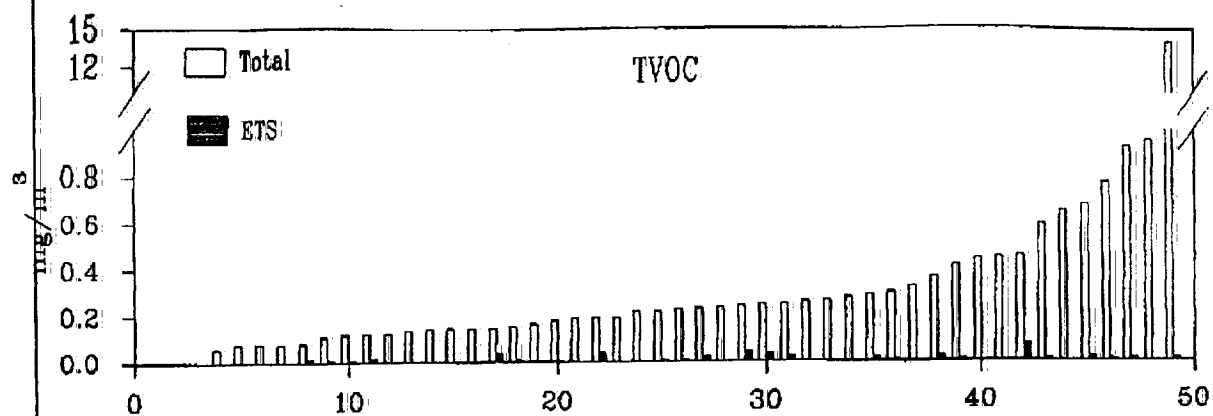
Home #	Apportionment %							
	TVOCs	Benzene	Styrene	Pyridine	2-Picoline	3-Picoline	4-Picoline	3-Ethylpyridine
1	0.59	3.35	2.52	5.84	15.65	60.23	12.76	ND
2	2.97	5.47	10.04	18.02	28.40	144.43	31.15	ND
3	5.48	25.73	23.92	39.65	78.60	134.88	105.72	80.93
7	1.07	2.04	1.58	12.65	21.74	150.58	ND	ND
8	20.87	39.03	32.96	41.66	88.52	123.10	ND	ND
9	8.29	26.24	24.74	39.18	73.88	94.77	85.25	65
10	0.96	4.17	4.58	5.62	18.78	34.75	6.96	8
12	0.00	0.00	0.00	0.00	ND	ND	ND	8
13	2.54	27.63	30.60	45.98	74.14	102.28	183.74	308
15	14.66	41.26	39.36	80.01	84.40	90.08	24.43	39
18	0.83	4.67	6.94	19.94	15.01	18.72	8.36	13
20	0.06	10.23	5.58	6.99	15.31	19.78	10.21	13
27	8.95	33.28	19.30	78.68	67.08	88.40	ND	8
31	19.74	29.38	18.19	58.41	68.26	106.18	ND	8
33	18.54	40.21	43.51	28.84	82.41	91.63	58.33	62
36	6.98	17.63	12.63	70.81	41.92	27.76	ND	ND
38	7.49	13.23	9.36	22.66	47.91	57.36	ND	ND
40	10.86	9.39	11.00	99.66	66.38	70.47	ND	ND
41	ND	5.11	43.82	120.72	65.61	113.25	ND	ND
42	1.58	9.17	5.02	302.13	ND	79.43	ND	46.1
43	5.32	33.64	26.56	108.34	70.56	88.54	45.02	8
48	23.78	45.33	44.04	53.54	102.48	113.76	ND	ND
49	0.00	0.00	0.00	0.00	ND	0.00	ND	ND
50	16.75	63.17	58.14	115.79	80.50	100.03	43.28	95.3
54	0.00	0.00	0.00	ND	ND	ND	ND	ND
Mean	7.51	19.57	19.01	57.30	57.50	83.06	51.27	58.4
S.D.	7.87	17.56	16.92	64.38	27.09	41.43	52.15	36.0
Median	5.48	13.23	12.63	40.66	67.08	90.08	37.22	62.0
Minimum	0.00	0.00	0.00	0.00	15.01	0.00	6.96	9.0
Maximum	23.78	63.17	58.14	302.13	102.48	150.58	183.74	108.1

ND = None Detected

2026224082

List of Figures

Figure 1. Histograms of the total and the ETS fraction of selected VOC analytes in all homes (smoking and nonsmoking). The TVOC results are ranked by increasing concentration, and the benzene, styrene and 3-picoline results are matched with their corresponding TVOC result.



2026224084